Removal of polycyclic aromatic hydrocarbons (PAHs) by different physicochemical methods: A mini-review

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Abstract

PAHs are persistent organic pollutants spread worldwide in our environment, including air, soil, and water. They are a large class of organic pollutants released mainly from anthropogenic sources, including automobiles and incomplete combustion of fossil fuels, power plants, and natural sources like forest fires and volcanic eruptions. In recent years, a variety of treatment methods have been used to degrade PAHs in the environment. This paper reviewed the most frequent physicochemical methods for PAHs removal, including solvent extraction/soil washing, oxidation, ozone, solidification, and supercritical fluid extraction. The findings showed that combining physicochemical methods can be an effective option for better cleanup of PAH from contaminated areas.

Keywords: Polycyclic aromatic hydrocarbons (PAHs), Contaminated Soil and water, physicochemical methods

1 Introduction

Polycyclic aromatic hydrocarbons (PAHs) are hydrocarbons having two or more fused aromatic rings, released from natural (like forest fires and volcanic eruption) as well as manufactured sources (like the burning of fossil fuel & wood, automobile emissions). Because of their high hydrophobicity, PAHs get adsorbed into the surface soil or marine sediments and strongly bind to organic matter. The adsorption onto sediments and high hydrophobicity prolong the life span of these contaminants in the environment. Thus, they are persistent priority pollutants and continue to last for a long time in the environment, causing severe damage to human health owing to their genotoxicity, mutagenicity, and carcinogenicity (Kalf et al., 1997; Borji et al., 2020; Mukhopadhyay et al., 2020; Nguyen et al., 2020; Pirsaheb et al., 2020). PAHs compounds were detected globally in soil, sludge, water, and other contamination sources.

PAHs are among the essential classes of aromatic compounds that have caused a serious concern due to their toxic effects on humankind and the ecosystem. Thus, they are considered a significant threat to ecosystem stability because of their persistent nature. PAHs are ubiquitous chemicals because they are widely identified in every part of the environment, from soil media, air mixtures, water sources, or their traces found in various food products (Onyemaechi et al., 2018). The PAHs with low molecular weight (naphthalene, phenanthrene, anthracene, and fluorene) have less carcinogenic potential. Furthermore, they do not persist in the environment for a more extended period because of their high volatility and slight solubility in water (Rabani et al., 2020). However, they pose risks to aquatic organisms such as fishes because of their bio-accumulative characteristic.

These compounds can also increase the food chain level employing biomagnification and subsequently cause toxic effects to human beings after consuming these organisms as food (Law et al., 2002; Sun et al., 2019). On the other hand, PAHs with high molecular weight (pyrene, Benz[a]anthracene, benzo[a]pyrene, chrysene, and indeno [1,2,3-cd] pyrene) are identified as persistent chemicals in the environment because of their low volatility and their high oxidative resistance (Stogiannidis & Laane, 2015). Due to the widespread sources and health risks posed by PAH pollution, there have been rising concerns about the public's need to be well-informed about their occurrence, levels, and possible health risks presented in different environmental media. Moreover, comprehensive data available on their occurrence, levels, and distribution will be essential to promote effective management of environmental pollutants such as PAHs.

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2 PAHs removal methods

There are several methods to remove PAHs from contaminated soils, waters, and atmosphere to mitigate the possible risk of PAHs on the environment and human health. They are categorized as physical, chemical, thermal, biological, and phytoremediation processes. However, most of these methods have several disadvantages, such as complicated operating procedures and high investment and maintenance costs (Lamichhane et al., 2016). Table 1 shows the existing methods of PAH removal from the environment.

Table 1: Existing PAHs removal methods from the air, soil, and water

Physical and chemical methods	Biological	Thermal	Phytoremediation
Natural attenuation Solvent extraction/soil washing Soil vapor extraction Solidification/stabilization Electrokinetic remediation Photolysis Adsorption	Biostimulation Bioaugmentation Bioremediation Bioslurping Bioventing	Thermal desorption Radiation frequency Heating	Rhizofiltration Phytoextraction Phytostabilisation Phytodegradation

2.1 physicochemical methods for PAHs remediation

There are several physical and chemical methods for remediation of PAHs from contaminated soil and water. In this section, the remediation techniques used frequently based on published papers in recent years are listed.

2.1.1 Oxidation

Chemical oxidation treatment of organic contaminants in wastewaters has received interest from researchers (Ameta & Ameta, 2018). The main advantages of this method are effectiveness and fast remediation ability compared to the other methods that need on/off-site excavation treatments, and the cost is greatly dependent on the chemical agents needed. In addition, this method is not influenced by the toxicity of the contaminated sources and can be considered a suitable remediation technique for PAHs contaminated from the soil.

The primary function of chemical oxidants is to inject liquid/gaseous chemicals to the ground, a simple operation with low operational costs. Moreover, this technique can degrade oil contaminants without forming toxic compounds into more biodegradable compounds (Lim et al., 2016). The rates of PAH oxidation depend on several factors, for instance: the structure of the compound, the temperature, the molecular weight, and the strength of the oxidizing agent (Abdel-Shafy & Mansour, 2016). Before applying any chemical oxidant, the oil-contaminated site must be evaluated for any ongoing natural reduction processes(Agarwal & Liu, 2015). The two common oxidants techniques to remediate PAH from polluted areas are ozone and Fenton techniques (Wang & Wang, 2021).

2.1.2 Fenton

Fenton oxidation is a feasible remediation technology for PAHs. Several studies have addressed the use of Fenton oxidation for PAHs removal from contaminated soils (Yap et al., 2011). The main challenge for the only Fenton treatment is the necessity of high oxidant load; however, integration of the Fenton technique with other treatment methods is developed to address only Fenton limitations. Meanwhile, deterioration of soil quality, PAHs unavailability, acidification of medium and microbial community are the major limitations. This technique shows promising potential for the fast Degradation of PAHs in the contaminated areas due to the utilization of hydroxyl radicals generated by the catalysis of hydrogen peroxide (Reddy & Chandhuri, 2009). Several chemical reagents could boost the PAHs availability in polluted soils to combine with Fenton oxidation, such as vegetable oil, co-solvent, cyclodextrin, and surfactant (Yang et al., 2017). For instance, co-solvents are used to increase the solvability of PAHs in the soils that eventually reduce the surface tension accurse between compounds in soil and the aqueous phase, which leads to an increase in the PAHs availability (Usman et al., 2016). Yap (Yap et al., 2012) suggested applying ethyl lactate-Fenton treatment for soils polluted with PAHs. This method resulted in 97% PAH degradation compared to the conventional Fenton oxidation method (31–57%). In another study, Lin (Lin et al., 2016) applied ultrasound and Fenton processes to the Degradation of PAHs in sludge. They found the degradation efficiency of PAHs in 30 min with the following order: Fenton (83.5%) >ultrasound–Fenton (75.5%) >ultrasound (45.5%). Recently, Mosmeri (Mosmeri et al., 2019) used CaO2 nanoparticles for decontamination of benzene-contaminated groundwater remediation by modified Fenton (MF) reaction through the continuous flow sand-packed plexiglass reactors. As reported, MF reaction showed a great potential of up to 93% in benzene removal in the first 20 days while the remediation was dramatically reduced to 75% at the end of the study.

2.1.3 Ozone (O_3)

O_3 is identified as one of the valuable chemical oxidation technologies for the Degradation of hydrocarbon from contaminated areas as it can be quickly delivered to the contaminated vadose zone (Lim et al., 2016). It should be noted that the presence of natural metal oxides such as goethite, Al_2O_3, and MnO on the soil's surface helps catalyze the decomposition of ozone to hydroxyl radicals (Lim et al., 2016). O3 is a strong oxidant and can quickly oxidize the organic compounds, reducing the remediation time and costs (O'Mahony et al., 2006; Krishnan et al., 2017); used this method to remove several types of phenanthrene from polluted soils. They obtained over 50% reduction in all types of phenanthrene concentrations after 6 hours of remediation. While Jonsson et al. (2006) reported that Fenton reagent was

more effective for PHAs remediation by 40–86% than O3 with an efficiency of 10–70%. Hence, using O3 for the Degradation of lower molecular weight PAHs was more effective than the oxidizing agent (Bavel, 2006).

2.1.4 Solvent extraction (SE) / Soil Washing (SW)

SE is a standard and viable employed remediation technology for removing PHAs from the polluted area, which could be applied lonely or mixed with other solvents (Gan et al., 2009). Meanwhile, SW is an ex-situ treatment technology used for various organic and inorganic polluted soils and involves high shear energy for the solid-liquid separation process. This technique is based on the desorption of PAHs from the binding site in/on the solid matrix through the action of simple water, organic solvents, or surfactants, followed by rinsing from the solid into the extraction fluid (Gan et al., 2009; Lau et al., 2010). In addition, the liquid phase contains desorption of PAHs, which causes further biochemical treatments to complete the detoxification process (Conte et al., 2005). For example, a study conducted by (Khodadoust et al., 2000) revealed over 95% PAH extraction when a solvent mixture of pentanol and ethanol was applied in three different stages.

2.1.5 Soil vapor extraction (SVE)

SVE removes harmful pollutants from the soil in the form of vapors (Speight, 2017). In this method, a negative pressure gradient should be applied to move vapors toward extraction wells. Volatile contaminants are gradually removed from the subsurface. Finally, the collected vapors are discharged into the atmosphere. SVE is applicable for both contaminants with the low and high volatility of organic compounds. Hence, it is not suitable for soils with low air permeability, as the induced air prefers to flow into lower resistance paths (Lim et al., 2016). The treatment of the downstream contaminant and sweep gas is the main disadvantage for SVE, which is costly for subsequent treatment downstream (Lim et al., 2016). Several researchers have used this technique for the removal of different PAHs from different contaminated sites. For instance, (Simpanen et al., 2017) found the effectiveness of SVE for the removal of BTEX from wet gasoline-contaminated soil. In another study, (Albergaria et al., 2012) used SVE to remediation artificially contaminated sandy soil with six types of PAHs. Based on their findings, 92% of removal efficiency obtained for contaminants with lower vapor pressures while longer remediation time is required. As found by (Soares et al., 2010), 92% of benzene removal from contaminated soil obtained using SVE.

2.1.6 Solidification/stabilization (S/S)

S/S is a remediation method based on physical changes and relies on the reaction between the reagent and the soil to reduce the contaminant's mobility (Hao et al., 2011). This method is occurring by a chemical reaction between solidifying reagents and waste by mechanical processes. Therefore, to reduce different environmental impacts, this method can be used in physical and chemical processes. In the stabilization, chemical reactions converting the pollutant in the less mobile species are interested, while in the solidification processes, the pollutants are physically linked to a solid matrix. (Ma et al., 2018) developed a novel method for PAHs-contaminated soil using 0.02% of sulfonated oil as the optimum dosage. They found that the PAHs leaching contents of remediated soil were 25% compared to the control sample.

2.1.7 Supercritical fluid extraction (SCFE)

SCFE is another proper and effective technique that uses fluid by heating and pressure above the critical point. SCFE works based on gaseous–liquid properties with high diffusivity and low viscosity due to the high temperature and pressure (Almeida et al., 2016). According to the previous studies, contaminants such as PAHs can be rapidly extracted from various environments. Additionally, based on factors like molecular masses and concentrations, flexibility in application, and aqueous solubility, SCFE is considered a feasible method for predicting the bioavailability of organic contaminants (Riding et al., 2013).

2.1.8 Electrokinetic remediation (EK)

EK remediation is an *in-situ* method for treating low permeable PAHs-contaminated sites (Gitipour et al., 2018). This method uses low levels of direct electric current between anodes and cathodes electrodes that impacted each side of the PAHs contaminated soil (Lim et al., 2016). It also should be stated that the application of this technique alone or in combination with other techniques such as Fenton or bioremediation is proven as a promising method for PAH removal.

The main challenge of using conventional EK remediation technology is slow desorption rates and hydrophobicity of PAHs from subsurface environments (Pazos et al., 2010a). So, it is essential to raise PAHs desorption from the polluted area and create a favorable environment to transport the electrode chambers to improve the EK process. Moreover, by increasing the electroosmotic flow, removal of an organic compound would increase with the presence in the processing fluid of electrolytes and pH control at the electrode chambers (T. Alcantara & 89–94.; Pazos et al., 2010b). The other advantage of this method is faster response time and lower operating costs as it can be treated in situ. Alcántara (Alcántara et al., 2010) carried out the EK remediation of kaolin contaminated with a mixture of different PAHs (fluoranthene, pyrene, and benzanthracene) and reported the high potential of this technique for remediation of polluted soil by 40%.

2.1.9 Thermal desorption (TD)

TD is a suitable technology for the Degradation of PAHs compounds with comparably high efficiency (Zhang et al., 2017). The primary mechanism of this method is the removal of volatile organic contaminants by using an outside heat source. Hence, the two main disadvantages of this technique are high operating costs and the potential of air contamination due to the desorption of organic compounds (Li et al., 2009). TD technologies are promising methods for removing hydrocarbon from polluted soils due to the rapid and reliable removal standards level. However, soil properties can be damaged because of high temperature and

energy consumption (Vidonish et al., 2016). The parameters that affect the efficiency of TD include temperature, soil composition, concentration and composition of contaminants treatment time, bulk density of soil, vapor pressure of contaminant, moisture content, and particle size distribution (Falciglia et al., 2011). An improvement in PAHs removal efficiency of 95% was achieved by applying TD (Bates et al., 2008). In another study, the TD was successfully applied to remediate PAHs in soil under different circumstances. They found that the removal efficiency reduced with decreasing residence time and heating temperature (Jeon). Table 2 shows the PAHs removal by different physicochemical methods.

Table 2: The removal of PAH from contaminated soil and water by different physicochemical methods

Location	Technique	Contaminated site	PAHs type	Removal rate (%)	Reference
France	Oxidation with Potassium permanganate	Soil	Acenaphthene, Acenaphthylene Pyrene, Fluoranthene Benzo [a] anthracene Chrysene, Benzo [a] pyrene	70	(Lemaire et al., 2013)
China	Ozone	Soil	Diesel	92	(Li et al., 2014)
Brazil	Photo-Fenton	Water	16 PAHs	92	(Rocha et al., 2018)
Malaysia	Electrochemical Process	Water	16 PAHs	95	(Yaqub et al., 2017)
France	Thermal pre-treatment and chemical oxidation	Soil	9 PAHs	30- 47	(Usman)
Argentina	Oxidation (permanganate, hydrogen peroxide, and persulfate)	Soil	phenanthrene and pyrene	63	(Peluffo et al., 2018)
France	Oxidation	Soil	16 PAHs	70	(Lemaire et al., 2013)
Norway	EK remediation	Soil	16 PAHs	35	(Pedersen et al., 2017)
Iran	SCFE	Soil	Naphthalene, phenanthrene, anthracene and pyrene	85	(Khanjari et al., 2016)
China	Stabilization/solidification	Soil	PAHs	25	(Ma et al., 2018)
China	Oxide/Ag_3PO_4	Water	Naphthalene, phenanthrene and pyrene	82-99	(Yang et al., 2018)
Tanzania	Washing	Soil	PAHs	35-99	(Mahugija & Njale, 2018)
France	Electro-Fenton treatment	Soil	Cenaphthene, phenanthrene, fluoranthene, pyrene benzo (a) pyrene, and benzo (g,h,i) perylene	99	(Mousset et al., 2016)
China	Washing followed by ozone oxidation	Soil	phenanthrene	45-99	(Liu, 2018)
China	Oxidation	Water	16 PAHs	45-82	(Qiao et al., 2017)
Mexico	Ozonation	Soil	phenanthrene	80	(Rodriguez et al., 2017)
United Arab Emirates	Electrochemical treatment	Water	phenolic compounds	100	(El-Ashtoukhy)

3 Future perspectives and Conclusion

PAHs are widely found in nature and accumulated, enriched, or produced in food through multiple paths, posing a significant risk to human health. The determination of PAHs still needs improvement. Previous efforts, including equipment and pretreatment, have made significant progress in PAH analysis types. Additionally, the development trends of future detection methods are mainly reflected in the miniaturization of instruments and equipment, the simplification of pretreatment processes, and the miniaturization of test samples. Moreover, due to PAHs' widely varying physical and chemical properties, their measurement is often complicated and costly. Methodology for sampling, analysis, and emission estimation will need to

be harmonized to adequately assess current ambient concentrations, the effect of future control measures and to refine any further action which may be required to assess their human health impacts adequately.

The following Conclusion can be obtained from the literature review: the most used physicochemical methods for PAHs removal were oxidation, ozone, and SCFE with 40 to 90% PAHs removal efficiency. Hence, only 20 to 50% PAHs removal efficiency was obtained using Stabilization/solidification and washing methods. Furthermore, the phenanthrene and benzene compounds were dominant PAHs in soil and only a few in water samples. Therefore, for future studies regarding PAHs removal from the soil, water, and air, we suggest:

1. Establish the new rules to reduce the emission of PAHs in the atmosphere of urban areas, especially in big cities with a high population.
2. Prevention of PAHs emission to the air by source identification in different study locations by implementing awareness programs for people.
3. Conduction of more research to investigate the combined Physico-chemical removal methods as they performed better than single methods.

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